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Next 2 Page(s) In Document Denied

25X1

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ESTIMATION OF ABSORBED

RADIATION IN VIVO

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We have occupied ourselves with this problem because we think that it should be solved today, with the rapid development of our industry in a period in which we are establishing many new productive processes. These processes may lead to various damages to our health, caused by the absorption of radioactive materials with a long half life, which were described in other countries in previous years. Further, the fact that we are a uranium-producing country, and the possibility of making use of atomic energy for peaceful and military purposes, have led us to work on this problem.

The medical profession itself noticed the seriousness of poisoning by radium, perhaps for the first time, upon the deaths of a group of people from the use of a "medical treatment by preparation of radiothorium" and various "curative" waters containing salts of natural radioactive elements with long half life. 1) 2) But Plum, 3) a New York dentist, had already aired in September, 1924 the suspicion that osteomyelitis of the

## Pracovni (2)

jawbone, which resembled phosphoric necrosis in persons employed in the painting of numerals with luminescent paint, was in fact formed by the effect of radioactive materials introduced into the mouth. Upon extraction of a tooth, the jawbone did not heal, and in the cases written up later this symptom was one of the first signs of poisoning by radium.

The problem of industrial poisoning, especially in the manufacture of luminescent numerals, was concisely stated in Martland's work in 1929. 4) It is based primarily on his experience with cases from a certain factory in New Jersey, where the manufacturing process included the painting of luminescent numerals on watches and various apparatus. In that factory, during the years 1917-1924, about 250 girls on the average worked in this production. Altogether during this period there were about 800 employed in the workshops of this factory. Among these girls there were fifteen cases of death suspected to be from poisoning by radium, and in five other cases poisoning by radium was shown to be the cause of death. Many other cases may have escaped observation because they died elsewhere and were diagnosed differently.

The luminescent paint used was composed of zinc sulfide with a mixture of radium, mesothorium and radiothorium in amount sufficient to provide satisfactory luminescence. The luminescent paint was prepared in such a way that small quantities of cadmium, copper and manganese were added to the repeatedly scorched zinc sulfide to improve luminescence. To this mixture was added a solution of radioactive salts which then were precipitated out

UNCLASSIFIED

Pracevni (3)

as undissolved sulfates. The color was suspended in a solution of acacia gum and diluted with distilled water. The workers then applied the paint to the numerals with a brush which they were accustomed to sharpen between their lips to obtain better results in their work. Martland estimated that an employee swallowed weekly from 15 to 215 gammas of radioactive materials.

The first deaths came in the years 1922, 1923 and 1924. Nine of the girl workers died after a period of work in the factory. They all died while being cared for at home, with these diagnoses (posthumous documents): ulcerative stomatitis, syphilis; primary anemia, Vincent's angina; poisoning by phosphorus, necrosis of the jawbone; industrial poisoning of undesignated type, necrosis of the jawbone; anemia, necrosis of the jawbone, etc. The corpse of one of the workers was exhumed five years later and high radioactivity was found in the bones.

Clinically Martland distinguished two types of cases, fulminant and chronic. The fulminant type is characterized by severe leucopenia, necrosis of one or both jawbones, with beech-nutlike lesions and often terminal sepsis. The first symptoms appeared from one to seven years after discontinuing the work of painting numerals. The author unfortunately did not state how long the exposure was for these women. According to Evans 7) this type manifests itself in persons having ten to one hundred gammas of radium in the body, and anemia then appears in the course of a few years.

Martland observed the chronic type for too short a time and therefore decided that necrosis of the jawbone is not the

CONFIDENTIAL

Pracovni (4)

characteristic symptom. For the same reason he did not observe changes in the blood. Evans 7) in his work in 1934 supplemented Martland's statement in this sense, that in these later cases he often comes upon changes in the blood, reporting in a few cases terminal necrosis of the jawbone and the sarcomatous bones. Inflammation of the bone had already manifested itself previously in various parts of the body. The content of radium in the body in these cases varied between one and ten gammas absolutely, and the symptoms of disease appeared in ten to fifteen years. This shows that it is necessary in these cases to supplement superficial clinical and hematological investigations with a determination of the quantity of absorbed radium, because we cannot expect that excessive quantities of absorbed radium will in time be manifested in clinically or hematologically identifiable disorders. It is possible for us to state that periodical examinations should regularly be made, that in the cases of these workers some change should have been observed, and that it might take years after the discontinuance of exposure before the appearance of these symptoms of poisoning by radium, which has unfortunate prognosis.

Table 1 shows the distribution of radium in the body in relative figures, taken from Evans' work. 5)

Fracovni (5)

Table I

	I	II	III
Vertebrae	100	100	100
Jawbone	-	51	20
Femur	-	48	27
Tibia	-	30	-
Skull	-	18	-
Rib	-	-	11
Teeth	-	-	46
Heart	0.5	-	0.04
Brain	-	0.4	-
Stomach	-	-	-
Liver	65	0.5	0.06
Intestines	2	-	-
Spleen	8.4	0.9	0.05
Lungs	17	2.2	0.04
Kidney	0.4	-	0.17

- I By intravenous injection  
 II Person who painted numerals with  
 luminescent paint  
 III Medicinal ingestion of radium per os

Especially striking is the relatively high concentration in the parenchymatous organs with intravenous injection. But this could not be characteristic on account of the manner of introduction; we think rather that it is caused by the large single dose and that death occurred relatively soon after injection. The cases shown under II and III absorbed radium over a long period of time, and the columns do not include the large part of the radium that was circulating, the remainder being deposited in the bones.

Barker and Schlundt 6) examined ten patients to whom medicinal radium chloride had been administered at various times. See Table II, page 6.

UNCLASSIFIED

Pracevni (6)

Table II

<u>Case</u>	<u>Diagnosis</u>	<u>I</u>	<u>II</u>	<u>III</u>	<u>IV</u>	<u>V</u>
I	Myocarditis, diabetes mellitus	1455		1455	1 day	4-8
II	Stomach ulcers	300	80	380	2.5 yr.	none
III	Polyarthritia and myocarditis	180	70	250	1.5 yr.	trace
IV	4 plus Wassermann, hypertension and glycosuria		890	890	20 mo.	19-33
V	Demencia praecox		80	80	7 mo.	none
VI	Phlebitis of the left leg, myocard- itis	120	180	240	5 yr.	none
VII	Myocarditis, hypotension		330	330	2 wk.	9-14
VIII	Cardiac asthma		305	305	5 wk.	9-14
IX	Urethral gonorrhea		1130	1130	20 mo.	10-18
X	Hypotension	92	115	207	2 yr.	none

Columns: I Ra given per oz. mg  
 II Ra given intravenously, mg  
 III Total Ra given mg  
 IV Time since latest treatment  
 V Ra remaining in organism mg

Patient I received radium for a period of 2.5 years, Patient II for a year, Patient III for a year, Patient IV for a year, Patient V for three months, Patient VI for four months, Patient VII for three years, Patient VIII for three years, Patient IX for three years, Patient X for one year. Negative values shown in the columns of Table II can be considered as due to too little of the method used. (Translator's note: No negative values

UNCLASSIFIED

Fracovni (7)

visible.) But this table demonstrates how small is the percentage of radium that is retained in the body, especially when taken orally. Evans 5) thinks that 60 to 98% of radium introduced into the organism is released again, of this about 90% by stool and 10% by urine. Not to distinguish the way of admission is certainly incorrect, as is shown by Table II. Radium injected into a vein is retained to a greater extent than radium entering the body by the digestive tract. It is evident that substantially more radium is retained in the respiratory part than in the digestive tract.

The "medicinal" administering of radium, whether per os or by injection, today has no significance. But certainly groups of our workers are either in fact or potentially endangered. Like the working women who painted murals with luminescent paints, so too are radiochemists and of course miners in uranium mines, of whom it can be expected that they inhale radioactive dust at their work, so that a dangerous amount of radium accumulates in their bodies.

However we do not know what to consider a dangerous dose of radium in the body. Aub and Evans 7) examined seven workers who had between 0.02 and 0.5 gammas absolute of radium in their bodies but who did not have symptoms of chronic poisoning by radium even after a period of seven to twenty-five years. On the other hand, Martland found fatal cases with a content of 1.2 to 2.0 absolute gammas. On the basis of these measurements a dose of 0.1 gammas absolute of radium in the entire body was



Pracovni (8)

established in 1941 (National Bureau of Standards) as the safe limit of tolerance. We do not know, of course, whether that dose is indisputably safe. Certainly the number of cases observed, of which Evans speaks, is too small and the observations too brief for us to be able to decide on the safety of this limit. Furthermore, that these people were not stricken with the classical form of poisoning by radium does not prove that their resistance to infection was not lowered, and that it is altogether safe to be the bearer of 0.10 gammas of radium. It is therefore up to our efforts, by suitable practicable hygienic working arrangements, and by instruction of all workers regarding the danger in handling radioactive materials, to limit exposure of the workers to the minimum. For sketches of safe working arrangements in the painting of numerals see 7) and 8).

We have also concerned ourselves from this viewpoint with the degree to which miners are in danger in uranium mines, as well as persons who have over a period of many years had exposure to immeasurably accumulated radium in the body. In contrast with Martland's cases, where the main place of admission was the digestive tract, but where the respiratory tract cannot be eliminated as a possible place of entry, in miners the respiratory tract is indicated as the entrance point for radioactive dust. Perhaps this difference can explain the differing clinical forms. We will not spend our time on the question of the gravity of this disease nor on the clinical form. For further information see 9) to 11).

Pracovní (9)

Behounek and Fort 12) measured the activity and extent of radium in the lungs and spine of ten affected miners and the activity and extent of radium in the lungs of eleven additional miners. From the results of these measurements the authors concluded that in the origin and development of ailments in Jáchymov the dispersing radiation is inhaled excretion, where long-lived radiation of deposited radioactive materials is not enough to explain the ailment. What we will show, on the basis of our measurements on living miners after long years of exposure, is that the total amount of radium introduced into the body is not enough for it to be a valid etiological factor in the origin of the disease in Jáchymov.

Determining radium in the body:

We can show this by several means:

1. Measurement of gamma radiation of disintegration products of radium in the body (radium B and radium C)
2. Measurement of radon in exhaled air
3. Estimation of radium in the urine and stools

This last method is not acceptable for establishing the quantity of absorbed radium in the body because the radium eliminated in the urine and stools depends much upon the absolute quantity of radium in the body, but also on the form into which it is combined in the body. Less radium deposited in the bones is eliminated in urine and stools than is radium temporarily deposited in other regions. Upon the introduction of radium

Pracovni (10)

into the body, whether into the respiratory or digestive parts, it will be eliminated to a great degree in a short time, but over a long period of absorption of radium a greater amount will be deposited in the bones and its elimination will therefore be substantially reduced. Because there is no possibility of accurately establishing the distribution of radium in the body in vivo, we cannot judge from the amount of radium in the urine or stools the entire amount of deposited radium.

The measurement of gamma radiation is the oldest method of determining absorbed radium in exposed workers. Schlundt, Barker and Blinn 13) made measurements with a quartz thread electrometer and were able to find five gammas absolute of radium in the body. Ives, Knowles and Eritten 14) worked with a double-threaded wulf electrometer placed at a table at which examinees were seated. They measured gamma radiation with an error of plus or minus about 0.3 gammas. The absolute quantity of radium in the body which they could measure also varied in whole gammas. Barker and Schlund 6) used the wulf-Hess electrometer. With use of the Geiger-Müller counter the sensitivity of measurement of gamma radiation was increased, and Braaten and Leitch 15) could find a dose of 0.25 gammas of radium. But with insensitive apparatus it is possible, as Hess and Niff showed in 1947 16), to show the minimal dose of 0.05 gammas.

The most ordinarily used method for the estimation of absorbed radium is based on the estimation of radon in exhaled air. It is known that radium breaks down into radon and then

UNCLASSIFIED

Pracovni (11)

further into radium A, radium B, radium C and so forth as far as radium G which is a stable unreactive isotope of lead.

One gram of radium produces per second  $2.097 \times 10^{-6}$  curies of radon. We see from the work of Evans 17) that about 45% of the radon originating in the body by the disintegration of radium goes into the exhaled breath, whereas 55% of radon thus originating is broken down into more stable disintegration products before it is exhaled by the lungs.

On this basis it is possible to judge from the concentration of radon in the exhaled breath the content of radium deposited in the body. Previously written works like Evans 17), Ives 14) and Barker 6), based on this principle, gave results that were reproducible to only a relatively poor degree. This was due partly to the rather low sensitivity of the apparatus--they used an ionization chamber and an electrometer--and a further source of error was disregard of the normal content of radon in the atmosphere in the laboratory, and naturally taking a sample of exhaled breath under these conditions is not acceptable. Harley, Jetter and Eisenbude 18) markedly improved this method by arranging that, in the taking of the sample of exhaled breath, the persons being examined breathed air from a cylinder that had been stored for a few weeks, so that the radon originally contained in the air had lived its life and the examinees therefore inhaled air free of radon. In this case the exhaled air contained radon originating from the disintegration of radium in the body. Measurement was carried out in an ionization chamber with an

CLASSIFIED

Pracovní (12)

impulse counter, and they found a concentration of  $10^{-14}$  curies of radon per liter of exhaled air, which corresponds to 0.001  $\mu$ gammas of radium in the body.

The authors, in their new work in 1951, did not eliminate errors arising from careless taking of the sample of exhaled breath, and therefore significant errors occur also in it.

In working out our method of measurement we have had two considerations, to remove the deficiencies of the methods used previously and to make these measurements with apparatus which was at our disposal. Originally we wanted to work, like Harley etc. 18), with an ionization chamber and an impulse counter. We got this equipment, however, only at the end of 1952, and up to now it has not satisfied our requirements for sensitivity and accuracy. We have for this reason worked with an Israel emanometer, made by the State Radiological Institute in Prague. As the sensitivity of this apparatus is not sufficient for finding small quantities of radon, with which it is necessary to begin in the measurement of persons exposed to dangerous absorption of radium, we concentrate radon from the larger volume of exhaled air by condensation with liquid air.

The Israel emanometer used by us is made up of two cylindrical brass chambers of the same size (inside diameter 2r is 30 cm, inside height v is 30 cm) with a volume of 21,200 cm<sup>3</sup>. Into the several chambers, which previously were evacuated, was let air containing emanations, the effects of alpha particles originating from the breakdown of radon, ionizing the air in

UNCLASSIFIED

Pracovní (13)

the chamber. With a voltage of 240 V between the collecting electrode and the jacket of the chamber an ionized charge collects in the electrode and in the jacket of a condenser of known size ( $C = 11.1$  plus or minus  $0.14$  cm), which is coupled with a single-thread electrometer. The growth of potential  $V$  in condenser  $C$  we can compensate for by voltage  $V$  from an auxiliary source and we can measure the amount of this compensatory voltage with an accurate voltmeter.

The principle of the measurement:

The alpha particle of disintegrating radon ionizes the air in the chamber. Under high enough voltage between the jacket of the chamber and the collecting electrode a completely ionized charge is collected in the electrode and in the condenser of capacity  $C$ ; a saturated current flows from the chamber. The magnitude of this current indicates the content of emanation in the chamber. If we see that the quantity  $1$  C of radon produces ionized current  $S$  of absolute electrostatic units, then the measured current  $i$  indicates that the amount of emanation in the chamber is:

$$E_m = \frac{1}{S} \quad (1)$$

$S$  is the equivalent in current; current  $i$  is given by the equation:

$$i = \frac{C}{300} \cdot \frac{V}{t}$$

UNCLASSIFIED

Pracovni (14)

$C$  = capacity of the condenser in centimeters,  $V$  is the growth of charge in volts in condenser  $C$  in a period of  $t$  seconds.

If we substitute in equation (1) from equation (2) we get:

$$E_m = \frac{C \cdot V}{300 \cdot t} \cdot \frac{1}{s} \quad (2)$$

In view of the natural lapse of time in the chamber, we write for  $t$ ,  $\frac{t' - t}{t' + t}$  thus:

$$E_m = \frac{t' - t}{t' + t} \cdot \frac{C \cdot V}{300} \cdot \frac{1}{s} \quad (3)$$

Because radon breaks down into further products, radium A, radium E, radium C, the current equivalent  $S$  sometimes increases, the value  $S$  for the single time  $T$  from the beginning is filled in the tables. Finally the proportions of the ionized chamber have the effect that several alpha particles strike the wall before they can shift all their energy into ionization. We measure then less ionization current  $i$  than would correspond to the full use of all the alpha particles. For the cylindrical ionization chamber there has been empirically estimated the relation which for our ionization chamber has this form:

$$i' = a \cdot i = (1 - 0.572 \cdot \frac{z_1}{r}) \cdot i = 0.8856i \quad (4)$$

where  $a = 0.8856$  is the correction for the size and form of the chamber.

In view of the markedly elementary recombinatory capacity of ions originating from ionization by alpha particles, and in view of the heterogeneous dispersion of electrical fields in

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UNCLASSIFIED

Pracovni (15)

the ionization chamber, in determining the voltage in the ionization chamber the current does not have to be a saturated current. Therefore there was introduced into the equation a correction factor in saturation  $c$ , which we determine for the establishment of measured current  $i$  from a graph.

The concentration of radon in the measured sample of air is then given by an equation which proceeds from equation (4), supplementing with single correction factors:

$$R_n = \frac{t' - 1}{t' + t} \cdot b \cdot n \cdot \frac{1}{s_T} \cdot \frac{1}{a} \cdot \left( 1 + \frac{c}{100} \right) \quad (5)$$

where  $t'$  = the period of time during which the thread measures  $n$ -units of work in measuring natural time lapse

$t$  = the period of time during which the thread measures  $n$  units of work, if the chamber is filled with air containing emanations

$b = \frac{C \cdot V}{300n} = 0.037 \cdot \frac{V}{n}$  = sensitivity to the charge

$n$  = number of parts in the scale

$S$  = current equivalent for the period of  $T$  minutes

$a$  = 0.8856 correction for the size and form of the chamber

$c$  = correction for saturation

Trial of the apparatus.

A comparative measurement was carried out. The same sample of radon was measured by electrometer with a small ionization

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Pracovní (16)

chamber, calibrated by the Oncological Institute, and measured  $1.42 \times 10^{-10}$  curies/liter, and then by our apparatus and was measured to be  $1.40 \times 10^{-10}$  curies/liter.

Further, a small concentration of radon was measured under laboratory conditions and at one atmosphere in a Karl box. The radon was concentrated from a few hundred liters of air (500-800 liters) condensed into liquid air. The measured concentration agreed with that given in the literature.

Concentrations found in the laboratory on various days:

$5.9 \times 10^{-13}$  curies of radon in one liter of air

$1.7 \times 10^{-13}$

$7.9 \times 10^{-14}$

$6.3 \times 10^{-14}$

In the Karl box there was measured  $7.2 \times 10^{-14}$  curies in one liter of air.

The radon content was further tested in air withdrawn from a cylinder of compressed air which was kept in storage about three weeks. 1,000 liters of air were concentrated and no radon was found.

As a further step we set about measuring the quantity of absorbed radium in unexposed persons and then in exposed persons.

#### Procedure of the experiment

Planned arrangement of the experiment - see sketch. The examinees breathed through a mask provided with intake and output vents air free of radon from a bag filled from a cylinder at least three weeks old. Between the cylinder and the bag a gas

Sketch of arrangement of the experiment:

Pracovni (17)

1. Cylinder of air
2. Gas regulator
3. Inhalation bag
4. Gas mask
5. Exhalation bag
6. Water cooler
7. Spiral chilled by ice
8. Cooled by solid  $\text{CO}_2$
9. U-tube chilled by ice
10. U-tube chilled by solid  $\text{CO}_2$
11. Tube containing soda-lime <sup>2</sup>
12. Tube containing calcium chloride
13. Flask containing phosphorus pentoxide
14. Copper spiral
15. Dewar vessel with liquid air
16. To air pump
17. Israel emanometer

UNCLASSIFIED

Pracovní (18)

regulator was installed. For about ten minutes the examinees inhaled air free of radon and exhaled freely into the laboratory atmosphere. In that time they breathed away radon held in the body which results, not from the breakdown of radium deposited in the body, but from an equilibrium between concentrations of radium in the external environment (in the laboratory) and concentrations in body fluids and tissues. By this time we could assume that practically all the radon exhaled does not come from outside environment but is produced directly in the body from a deposit of radium. 23) We connect the exhalation vents of the masks to an exhalation bag. From this bag we pump away the air by means of an air pump through a system of arrangements for ridding the air of moisture and carbonic acid and through a spiral tubing immersed in liquid air, in which we condense the radon. Drying the air and ridding it of carbonic acid is done as follows: We first of all conduct it through a water cooler, then through a spiral copper tubing chilled by ice, then through a pair of U-tubes immersed in ice, then a pair of U-tubes immersed in solid  $\text{CO}_2$ , then tubes containing lime-soda and calcium chloride, arranged in such a way that they can be changed without interrupting the experiment, and of course we conduct the air through a vessel over phosphorus pentoxide. From this vessel air comes off that is sufficiently dry and free of carbonic acid so that cooling in the temperature of liquid air does not precipitate water and carbonic acid. This is very important because it would be inadvisable for the preparation in the chilled spiral to

UNCLASSIFIED

Pracovni (19)

be frozen by the liquid air. We measure the speed of suction with this system by means of an attached flow meter.

We allow the examinees to breathe for about one hundred minutes and make a note of the exact period of time. In the course of this period of time we measure the free path of the ionization chambers which were evacuated and filled with air free of radon, with air brought through dry calcium chloride and free from ions of water glass. We take 15 to 20 measurements of the average value of  $t'$  (free path).

In measuring the free path and in completing the experiment on the examinees, we couple the spiral with condensed radon to the evacuated chamber and at normal temperature pump the radon into the chamber. The spiral having warmed up toward normal room temperature, we wash it with air free of radon and pump this air into the chamber. After a time, not more than half an hour, we begin with the measurement with which we establish the time  $t$ . Upon completion of the measurement we remove the emanations from the ionization chamber by means of repeated evacuation and washing with air free of radon. We again established from twenty measurements the average value of free path,  $t'_2$ . By adding the value  $t'$  (the average of  $t'_1$  and  $t'_2$ ) and  $t$ , and sensitivity of the charge  $b$  to the equation 3) we establish the quantity of radon in curies in the exhaled air. This quantity corresponds, as has been said previously, to 45% of the radon produced by radium deposited in the body. By converting this to 100%, divided by the entire time of breathing in seconds, and divided by the value  $2.097 \times 10^{-6}$  (the corresponding quantity of radon

ORIGINAL

Tracovni (20)

in curies produced by one gram of radium per second, we arrive at the quantity of radium deposited in the body, in grams.

This method of measurement has been used on a group of unexposed persons, some of whom were measured twice, some three times. We show the results in Table III.

Table III

<u>Person</u>	<u>Number of Measurements</u>	<u>Results in <math>\mu</math> Ra absolute</u>
A	1	0
B	1	0
C	3	4.8 x 10 <sup>-9</sup>
		7.4 x 10 <sup>-9</sup>
		4.3 x 10 <sup>-9</sup>
D	2	5.5 x 10 <sup>-9</sup>
		7.0 x 10 <sup>-9</sup>
E	3	7.2 x 10 <sup>-9</sup>
		4.8 x 10 <sup>-9</sup>
		2.0 x 10 <sup>-9</sup>
T	2	7.0 x 10 <sup>-9</sup>
		9.1 x 10 <sup>-9</sup>
D	1	4.9 x 10 <sup>-9</sup>
H	1	4.5 x 10 <sup>-9</sup>
CH	1	4.3 x 10 <sup>-9</sup>
I	1	0

We see that in almost all persons examined there were found varying amounts of absorbed radium in thousandths of grams.

We have found in the literature two works concerned with the measurement of deposited radium in unexposed persons. There is the work of Krebs from the Institute of Professor Rajewski in 1943 (19) and the work of Hirsch and Gates in 1950 (20). Both authors, Krebs in Germany and Hirsch and Gates in America, established the content of radium in the ashes of unexposed dead. They ground and separated the ashes, they watched for a

Pracovni (21)

state of equilibrium and then established the content of radium emanometrically. Table 3a gives the results of Krebs, Hursh and our own.

Table IIIa

<u>Authors</u>	<u>Number of Measurements</u>	<u>Average Amount</u>	<u>—</u>
Krebs	18	$13.89 \times 10^{-9}$ g	10.0
Hursh etc.	25	$0.159 \times 10^{-9}$ g	0.115
Müller etc.	16	$4.55 \times 10^{-9}$ g	8.57

The table shows us that Krebs obtained results from his material about two degrees higher than Hursh from his American material. To repeat, both authors worked with the ashes of unexposed deceased persons using similar methods. Our results with the living unexposed are statistically significantly higher than the American figures and significantly lower than the German. It seems to us almost verisimilar that these differences are due to different geological structures of the regions in which these persons who were examined lived. It would of course be necessary to support this hypothesis with knowledge and comparative work with the waters of those regions. We should also be able to study the radium content of the foods in those areas, because from the work of Evans and his associates (21) we know that some plants are capable--it has often been counted--of concentrating radium from water up to one hundred times.

Further, a radiochemist was then examined, in whose case it was possible to presuppose greater exposure, and these values

UNCLASSIFIED

Pracovní (22)

were found:  $2.8 \times 10^{-8}$  g  
 $4.7 \times 10^{-8}$  g  
 $3.5 \times 10^{-8}$  g

The amounts found in the examined radiochemist were quite higher than in the group of unexposed persons, but this worker did not exceed, according to our measurements, the maximal admission limit of 0.1 grams, although that amount is approached.

We have further examined a group of Jáchymov miners, all with long exposure underground. The values found were:

Table IVPatient Nr. 26851/52

Age: 51  
 Exposure: 21 years in uranium mines of which three to four were as a driver and timberman, the remaining time as an ore-crusher, now pensioned.  
 Clinical Diagnosis: The fibrocasseosa pulm. utr. Cavitaria dx. Calc. ossis calcanei bilat.  
 Measurements: (2)  $7.7 \times 10^{-9}$  g Ra absolute  
 $1.4 \times 10^{-8}$  g

Patient Nr. 27565/52

Age: 49  
 Exposure: 12 years of work underground in uranium mines  
 Clinical Diagnosis: Bronchitis chron., emphysema pulm.  
 Measurements: (1)  $9.5 \times 10^{-9}$  g Ra absolute

Patient Nr. 27871/52

Age: 52  
 Exposure: 14 years of work underground in uranium mines  
 Clinical Diagnosis: Bronchitis chronica. Emphysema pulmonum.  
 Measurements: (1)  $6.8 \times 10^{-9}$  g Ra absolute

Patient Nr. 28822/52

Age: 42

REF ID: A66117

Pracovní (23)

Exposure: 14 years of work underground in uranium mines  
Clinical Diagnosis: Bronchitis chronica  
Measurements: (1)  $7.1 \times 10^{-9}$  g Ra absolute

Patient Nr. 28412/52

Age: 45  
Exposure: 14 years of work underground in uranium mines  
Clinical Diagnosis: Bronchitis chronica  
Measurements: (2)  $5.9 \times 10^{-9}$  g Ra absolute  
 $5.6 \times 10^{-9}$  g Ra absolute

Patient Nr. 29960/52

Age: 53  
Exposure: 35 years of work underground in uranium mines  
Clinical Diagnosis: The fibrosa inact. apicis pulm. sin.  
Emphysema pulmonum.  
Measurements: (2)  $6.1 \times 10^{-9}$  g Ra absolute  
 $5.5 \times 10^{-9}$  g Ra absolute

Patient Nr. 30254/52

Age: 43  
Exposure: 13 years of work underground in uranium mines  
Clinical Diagnosis: Bronchitis chronica.  
Measurements: (1)  $3.2 \times 10^{-9}$  g Ra absolute

Average:  $7.14 \times 10^{-9} \pm 2.7$

In three of the last measurements the ascertainment of radon in exhaled air was measured simultaneously in gammae with a Geiger-Müller counter placed behind the examinee, very close to and parallel with the spine. In these measurements no significant difference was found between the value of the free path and our measurement. The apparatus made little distinction between such insignificant quantities of gamma radiation, as we anticipated.

It is interesting to compare with these figures the results of the work of Běhounek and Fort 12). These authors found in

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Pracovni (24)

the intact tissue of dead miners  $226-534 \times 10^{-14}$  g Ra in 1 g of spine and  $2.1-15.0 \times 10^{-14}$  g Ra in 1 g of fresh lung.

If we start out with the average weight of the spine as 1,300 g and the average value  $375 \times 10^{-14}$  g Ra in 1 g of spine, we get the value  $375 \times 10^{-14} \times 1,300 = 4.87 \times 10^{-9}$ . For the lungs, using the same calculation we arrive at the value  $0.06 \times 10^{-9}$  g absolute. We have neglected this value. If we suppose further that 80% of the radium is deposited in the spine, we arrive at a result of  $6.09 \times 10^{-9}$  g Ra absolute in the whole body. This value agrees well with our results.

#### Discussion

A method was worked out for determining the quantity of absorbed radium in vivo. The method we have used excludes the deficiencies of formerly used methods, primarily in taking the sample of exhaled breath. Since radon is present in the whole quantity of exhaled air (for example, over a 100 minute period), an error is avoided which arises in the taking of small samples, namely that a small sample of air breathed for example into an evacuated cylinder is an undetermined mixture of air from the miner's respiratory tract and alveolar air. If we assume that the air inhaled is free of radon, the effect of this mixture of alveolar air and air from the respiratory tract is a lowering of the measured values. This would not be especially objectionable if the error were constant. But we cannot assume this because there will always be retained in the cylinder some fractions of exhaled air. In our method this error is avoided

Pracovní (25)

because we concentrate the radon from all the exhaled air. Also, differences in the extent of ventilation cannot distort our results, because we did not calculate the content of radon in a unit of volume of exhaled air, but the entire quantity of radon exhaled in a unit of time. It therefore becomes a matter of complete indifference whether the quantity is breathed into one or two liters of air. A given quantity of radium produces in a unit of time a quantity of radon according to the disintegration constant (1 g of radium produces per second  $2.097 \times 10^6$  curies of radon).

The method we have described also makes it possible to determine the quantity of absorbed radium with sufficient sensitivity with apparatus which was at our disposal.

From our point of view the results of our measurement of the Jáchymov miners were quite important. They showed us that the Jáchymov miners do not absorb and do not retain in the body substantially more radium than normal unexposed workers. This result is at first sight quite surprising. But we remember the work of Behounek and Fort, who also did not find substantially greater radium content in the lungs and spine of deceased Jáchymov miners, and finally it can be assumed--since we do not have very exact information about the technology of work in uranium mines--that they do not directly drill uranium ore, since there would then be produced a pronounced dustiness of long lived radiation. According to the work of Behounek and Santholzer 22), the kind of rock that lies near uranium veins does not

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Pracovní (26)

have especially high activity. This fact may explain our finding.

But this finding has even more serious consequences. It is impossible to show either uranium or radium as the etiological factor of the Jachymov ailment. It will be necessary to think first of all of radon or of short-lived deposits of disintegration products of radon on inactive dust. We ourselves think that particles of dust bearing stable disintegration products of radon are a substantially more important etiological factor than is radon itself. This question will be discussed more fully in another work.

The method worked out by us is useful in ascertaining the degrees of danger in a worker with open radium apparatus, as in the application of luminescent colors, in radiochemistry and other similar situations where persons are endangered by the absorption of abnormal quantities of radioactive materials from the uranium series, higher than radon in order of disintegration.

Professor Herčík and Dr. Schober criticized this work in the seminar of the Institute and the Clinic for Occupational Diseases, as opponents, October 1, 1952.

The opponents showed in their report that it could not be correctly assumed, purely on the basis of the Evans work, that 45% of the radon produced from radium deposited in the organism is expelled in exhaled air. In the discussion it was granted that the selected way of estimating absorbed radium in vivo is possible only now, since previously the apparatus for measuring gamma radiation has not been accurate enough. For practical purposes no substantial objection was raised. In the International Congress of Roentgenology and Radiology in London in 1950, in setting the maximal admissible dose of absorbed radium, it was given out from Evans' statement and determined partly on the basis of 0.1 gamma of absolute radium, partly on  $10^{-12}$  curies of radon per liter of exhaled air, which corresponds to 45% of the radon produced by 0.1 gamma of radium.

In closing we thank our opponents and those who took part in the discussion for their comments.

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Pracovní (27)

Abstract

## Estimation of Absorbed Radium in Vivo

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A method was worked out for the estimation of absorbed radium in vivo. The method described successfully obviates the shortcomings of the methods hitherto described and makes use of home-produced apparatuses only. Apart from estimating radon in the normal laboratory atmosphere and in the city atmosphere of Prague, the amount of absorbed radium was estimated in an unexposed group, further in an exposed radiochemist and in a group of miners in Jachymov. The findings in the group of unexposed subjects are compared with the data in the literature, in the exposed radiochemist the figures obtained were significantly higher and in the group of Jachymov miners the figures found did not significantly differ from those found amongst the unexposed. In conclusion the significance of these findings is discussed.

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